EVALUATION OF PERFLUOROALKYL ACIDS (PFAAs) IN WATER ENVIRONMENT, FOOD, AND HUMAN BODY IN KLANG VALLEY, MALAYSIA(Abstract_要旨)

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論文題目	EVALUATION OF PERFLUOROALKYL ACIDS (PFAAs) IN WATER ENVIRONMENT, FOOD, AND HUMAN BODY IN KLANG VALLEY, MALAYSIA（マレーシア、クラン渓谷における水環境、食品、人体中ペルフルオロアルキル酸（PFAAs）の評価）		

（論文内容の要旨）

This thesis presents in-depth information on the level of Perfluoroalkyl acids (PFAAs) contamination in Malaysia, particularly in Klang Valley area. The thesis was divided into 8 chapters with each of the chapter covering various part as followed:

Chapter 1 Introduction

This chapter briefly explained the background and purpose of this research. The objectives, together with the scope of the studies were also covered in detail in this chapter.

Chapter 2 Literature review

Literature review mainly focused on the collection of literature, and the topics relevant to the research were covered. It started with the introduction of PFAAs including characteristic, usage; and followed by the concern on PFAAs to the environmental and human health. Most of the previous studies done on the analysis of PFAAs in environmental media and human were also covered in this chapter. The literature on the pathway to human exposure was also collected and presented in this chapter.

Chapter 3 Level and determinants of PFAAs in serum samples of the population in Klang Valley, Malaysia

This chapter focused on the level and exposure assessment of PFAAs in serum. The main objective of this chapter was to find out the significant determinants of PFAAs in the blood of the Malaysian population based on the self-report questionnaire. The types of determinants assessed were dietary intake, lifestyle, and socioeconomics. The participants of 219 from Klang Valley area were selected for this study. The analytical results showed that PFAAs concentrations up to 32.57 ng/mL were detected in the serum samples. At least 7 PFAAs were detected in serum samples of 78% participant with Perfluorooctane sulfonate (PFOS) being the predominant PFAAs (median = 8.79 ng/mL). Most PFAAs concentrations were higher in male and positively correlated with age, while BMI and smoking were not significantly associated with serum PFAAs. Among lifestyle variables, leather sofa and non-stick cookware were significantly associated with an increased level of some PFAAs. However, dietary intake does not appear to be the major determinants of PFAAs exposure in this population.

Chapter 4 PFAAs in surface water and sediment of Selangor and Langat river basins: spatiotemporal, partitioning, and ecological risk assessment

This chapter focused on the analysis of PFAAs in both surface water and sediment collected from

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Selangor and Langat River basins in the year of 2017. Spatiotemporal analysis together with the sediment-water partition was also assessed in this chapter. The presence of PFAAs in surface water and sediment of the Selangor and Langat River basins were quantified using LC-MS/MS. At least 3 and 1 PFAAs were detected in surface water and sediment samples, respectively. The total concentrations of PFAAs (∑PFAAs) in surface water samples in Selangor and Langat River basins were respectively in the range of 0.89 – 133.69 and 0.42 – 138.95 ng/L, while in the sediment the range of not detected to 249.7 and 18.7 – 465.6 pg/g dw were obtained. Perfluorooctanoic acid (PFOA) and Perfluorobutyric acid (PFBA) were found to be dominant PFAA in both water and sediment samples. The mean concentrations of PFAAs in sediments were higher than those in surface water. Urbanization and industrialization were identified as potential sources of PFAAs contamination in the area of Klang Valley. The logarithmic values of field-based K_D (partition coefficient) were in the range of 1.5 to 2.3, being slightly different from the previously reported values, due to the differences in the physicochemical properties of the environment. None of the PFAAs levels exceeded the PNEC value, suggesting little and/or no potential risk to the aquatic organisms.

Chapter 5 Levels and source exposure assessment of PFAAs in selected foods and drinking water collected from market within Klang Valley, Malaysia

This research revealed the first evidence on the contamination of PFAAs in selected food and drinking water samples from Malaysia, particularly in Klang Valley. Nine PFAAs were analyzed in 56 foods samples of 19 different types and 36 drinking water samples from the tap and bottled water using LC-MS/MS. In this study, PFAAs were found above the detection limit in all food samples and some drinking water samples. The highest level of mean ∑PFAAs in foods was found in chicken (5.78 ng/g ww), followed by beef (3.14 ng/g ww) and marine fish (0.93 ng/g ww). Overall, all marine food samples were the main contributor of long-chain PFAAs (C≥8), except for mollusk. Mollusk was the major contributor for PFOA while the other foods were the major contributor for PFOS. Tap and bottled water samples showed a varied concentration of PFAAs, which was dependent on the sources of their raw water. Based on the dietary intake exposure assessment, it was found that frequent consumption of chicken may lead to the health risk associated with PFAAs to the local population, due to the high presence of PFOS in chicken samples. Intake of other foods and drinking water will not cause any immediate health risk to human. However, health quotient above unity were observed for consumption of marine fish for adolescent, after taking into account the combination hazard ratio of PFOA and PFOS.

Chapter 6 Conclusions and future recommendation

This chapter summarized the conclusions of the important findings and their implications, as well as the recommendations for further research.

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本論文は、マレーシア、クラン渓谷を対象として、Perfluoroalkyl acids (PFAAs)による環境汚染やその人体負荷の実態を調査した結果を報告したものである。得られた主な成果は以下のとおりである。

1）　クラン渓谷の住民 219 人の血清中 PFAAs 濃度を測定した結果、その最大値は 32.57 ng/mL であり、78%のサンプルから 7 種の PFAAs が検出され、最も高濃度で検出されたのは Perfluorooctane sulfonate (PFOS)であった。ほとんどの PFAAs 濃度は男性の方が高く、また年齢との間に正の相関が見られたが、BMI や喫煙との間には有意な相関は見られなかった。また、革製ソファやテフロン加工調理器具を使用していることと、いくつかの PFAAs 濃度増加との関係が認められた。しかし、食品摂取が PFAAs 曝露の主たる経路とは考えられなかった。

2）　セランゴー川とランガット川における 2017 年の表流水中全 PFAAs 濃度はそれぞれ 0.89-133.69 ng/L と 0.42-138.95 ng/L だったが、底質中濃度はそれぞれ ND-249.7 pg/g-dw と 18.7-465.6 pg/g-dw であり、平均して底質中 PFAAs 濃度の方が高かった。また、濃度の空間分布から都市化や工業化がこの地域における PFAAs 汚染源と推定された。実測値から底質-水分配係数 K_D (L/kg-dw)を推定した結果、Log K_D 値は 1.5 から 2.3 程度であり、従来の報告値とは若干異なっていた。実測された PFAAs の中で、無影響濃度予測値（PNEC）を超えるレベルのものはなく、水系生態系へ及ぼす影響はほとんどないと考えられた。

3）　クラン渓谷の市場における食物 56 サンプル、飲料水 36 サンプル中の PFAAs 濃度を測定した結果、全食物サンプルといくつかの飲料水サンプルから PFAAs が検出された。平均全 PFAAs 濃度の最大値は鶏肉（5.78 ng/g-ww）であり、次が牛肉（3.14 ng/g-ww）、そして魚（0.93 ng/g-ww）であった。総じて全ての海産物は長鎖 PFAAs(C≥8)の主たる寄与源と考えられたが、軟体動物だけは例外であった。経口摂取曝露アセスメントの結果、鶏肉中 PFOS 濃度が高いため、全ての調査グループにおいて、頻繁な鶏肉の摂取は PFAAs による健康リスクを生じる可能性が示された。他の食物や飲料水の摂取が、すぐに人の健康リスクを生じることはないと考えられた。

以上の結果は、マレーシアにおける人と環境の PFAAs 汚染実態を示す貴重なデータであり、マレーシアにおける今後の PFAAs 利用計画の策定に大きく貢献するものであって、学術上、実際上寄与するところが少なくない。よって、本論文は博士（工学）の学位論文として価値あるものと認める。また、令和元年８月２１日、論文内容とそれに関連した事項について試問を行って、申請者が博士後期課程学位取得基準を満たしていることを確認し、合格と認めた。